

INTRODUCTION

Accurate parameterization of sea salt emissions in Chemical Transport Models (CTM) is important because naturally occurring aerosols contribute to ambient concentrations of particulate matter (PM). Natural and anthropogenic aerosols and gases interact chemically and physically, so predicting natural emissions correctly will affect the overall accuracy of the CTM. **AURAMS** (A Unified (multiple-pollutant) size- and chemical composition-resolved, episodic, Regional Air-quality Modelling System) is an off-line CTM driven by the regional Canadian operational weather forecast model, **GEM** (Global Environmental Multiscale). The **Canadian Aerosol Module (CAM)** within AURAMS is a size-segregated multi-component sectional aerosol algorithm dealing with major aerosol processes in the atmosphere including generation of sea salt aerosols. Using a sectional approach, the aerosol size distribution is approximated by a set of 12 contiguous, non-overlapping and discrete particle size bins, handling aerodynamic diameters ranging from 0.01 to 41 μm . The sea salt concentration in the surface model layer is given by:

$$c = b_e U_{10}^{1.0}$$

where U_{10} is the 10 metre wind speed in m s^{-1} . Coefficients a and b were found to vary by location in the North Atlantic where measurements were taken. The dependence of c on wind speed is weaker for smaller particles. Various parameterizations for sea salt flux are available in the literature. The default number flux function in AURAMS v1.3.1 is based on Monahan *et al.* (1986) for open ocean sea salt droplet generation through indirect bubble mechanism per unit area of sea surface as a function of 10 m wind speed in model grid squares with ocean fraction > 0.3. While wave breaking in the surf zone might be expected to generate more sea salt aerosol than over the open ocean, it is excluded in the default case. The results of testing two other open ocean parameterizations, plus one parameterization for the open ocean (Smith & Harrison) in combination with DeLeeuw or Monahan in an assumed surf zone, are shown here.

Sable Island

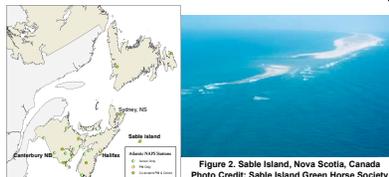


Figure 1. Map showing Atlantic Canadian Sites

Sable Island, a 44 km long sand bar 300 km southeast of Halifax, Nova Scotia, is ideally located to observe long-range transport of pollution aerosols from the northeastern US as well as background or natural sea salt aerosols in clean marine air masses. Aerosols arriving at Sable Island vary seasonally. Summertime weather is strongly influenced by high pressure systems over the western Atlantic which circulate air from the northeastern US to Atlantic Canada. Typically, aerosols during the summer are relatively small and dark, consisting of mostly sulphate with small amounts of soot. Speciation results at Sable Island indicated approximately one third of annual $\text{PM}_{2.5}$ mass is attributable to Sodium Chloride (Figure 3).

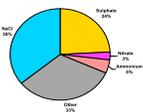


Figure 3. Sable Island (NAPS Station No. 31001) Partisol $\text{PM}_{2.5}$ Sampler Annual Average Speciation Analysis (November 2006 to September 2007, N=44)

Sea Salt Flux Parameterization Sensitivity in the CTM AURAMS

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Flux Parameterization Tests

February 2005

In February 2005, high $\text{PM}_{2.5}$ concentrations were measured at Sable Island. Often high $\text{PM}_{2.5}$ concentrations measured at Sable Island correlate well with wind speed, while at inland sites there is more likely to be a negative correlation with wind speed due to reduced mixing or stagnation resulting in locally higher $\text{PM}_{2.5}$ levels. $\text{PM}_{2.5}$ showed good correlation with wind speed at Sable Island in this case (Figure 4), while inland sites such as Canterbury, NB, showed a negative correlation (Figure 5). AURAMS predicted that almost all (99.8%) of $\text{PM}_{2.5}$ mass at Sable Island was fine sea salt. This is reasonable given the meteorological situation leading up to the event: persistent and strong northeasterly winds from a non-anthropogenic oceanic source region. The Monahan sea salt flux in CAM was found to over-estimate sub-micron sea salt aerosol, in the lowest model level (Figure 7). Other sea salt flux parameterizations were tested on this case, including two hybrids, whereby Smith and Harrison was used in grid cells with total ocean coverage (fraction of ocean = 1.0) and Monahan's or DeLeeuw's functions were called in grid cells containing some land fraction (fraction of ocean < 1.0), i.e., in an assumed "surf zone" around Sable Island (termed DeLeeuw SURF and Monahan SURF shown in Figure 7).

July 2007

Monahan's sea salt flux parameterization in AURAMS is suspected of over-predicting the proportion of surface fine sea salt aerosol at Halifax, NS, on 14 July 2007 (Figure 8). Using the hybrid parameterization of Smith and Harrison over open ocean grid cells (Ocean Fraction = 1.0) and Monahan in the assumed "Surf Zone" near the coast (in model grid cells with Ocean Fraction < 1.0), the fraction of fine sea salt aerosol was reduced (Figure 9), but the total $\text{PM}_{2.5}$ mass at Halifax was under-forecast in both cases.

Figure 10. Dalhousie Lidar Backscatter Ratio Measurements at Halifax, NS, 141400 -150000 UTC July 2007

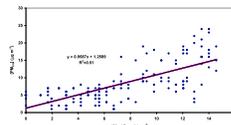
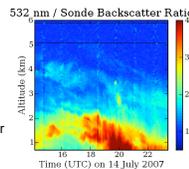


Figure 4. Measured $\text{PM}_{2.5}$ versus Hourly Wind Speed at Sable Island, NS 5-10 Feb 2005

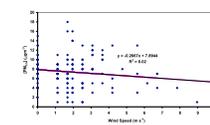


Figure 5. Measured $\text{PM}_{2.5}$ versus Hourly Wind Speed at Canterbury, NB 5-10 Feb 2005

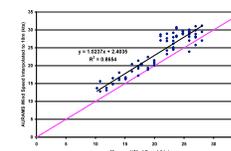


Figure 6. AURAMS Model Wind Speed Bias at Sable Island, NS, 5-10 Feb, 2005

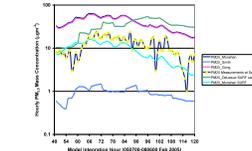


Figure 7. Comparison of model surface $\text{PM}_{2.5}$ predictions at Sable Island 5-8 Feb 2005 using Various Sea Salt Flux Parameterizations

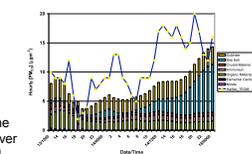


Figure 8. AURAMS Model Speciation of $\text{PM}_{2.5}$ at Halifax, NS, using Monahan's Default Sea Salt Flux Parameterization 13-14 July 2007

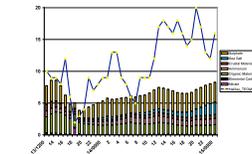


Figure 9. AURAMS Model Speciation of $\text{PM}_{2.5}$ using the "Monahan Surf" Hybrid Parameterization 13-14 July 2007

CONCLUSIONS

The Monahan sea salt flux in AURAM's CAM module was found to over-estimate predicted sub-micron sea salt aerosol in the lowest model level. While a "Monahan Surf" hybrid parameterization showed the best correlation with surface $\text{PM}_{2.5}$ measurements at Sable Island in one case from the winter of 2005, it did not show as much promise for cases in other seasons. Besides, Monahan's function was developed for the open ocean and not intended for use in the surf zone. Comparisons of several parameterizations for sea salt flux in the CTM AURAMS showed that none produced a satisfactory representation of fine particulate mass concentration at Atlantic Canadian sites during events when high mass concentrations were attributable to sea salt aerosol. In addition to finding a better modelled sea salt emission flux over the open ocean, it is desirable to include a parameterization developed specifically for the surf zone. Further study of seasonal correlations of wind speed, direction, particle mass, speciation and size distribution is needed to validate model predictions for Atlantic Canada. Sable Island is ideally located to study sea salt aerosol generated in the surf zone, with little interference from anthropogenic sources of Na or Cl, but additional instrumentation for speciation and size distribution is required. Existing instrumentation at Halifax, NS, can be utilized in determining a representative sea salt aerosol mass for coastal sites in the validation of Canadian CTMs.

$\text{PM}_{2.5}$ Composition

July 2007

The amount of sea salt that could be expected in summertime $\text{PM}_{2.5}$ at a coastal site such as Halifax, as indicated by the fraction of NaCl in 24 hour filter samples averaged over 6 sampling days in July 2007, is about 1% of reconstructed $\text{PM}_{2.5}$ mass (Figure 11). Using the "Monahan Surf" hybrid parameterization, AURAMS predicted, on average, 8% of $\text{PM}_{2.5}$ on 13-14 July was fine sea salt aerosol (Figure 12).

The analysis depicted in Figure 11 is missing Silicon, a large component of crustal material, as well as estimated particle-bound water (~13%). This could at least partially explain the high percentage (28%) of Unaccounted $\text{PM}_{2.5}$ mass in Figure 11.

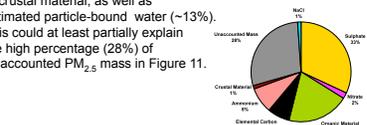


Figure 11. Halifax (NAPS Station No. 30113) $\text{PM}_{2.5}$ Sampler Average Speciation Analysis (July 2007, N=6)

Figure 12. AURAMS Model Speciation of $\text{PM}_{2.5}$ using the "Monahan Surf" Hybrid Parameterization 13-14 July 2007

Wet Removal

November 2007

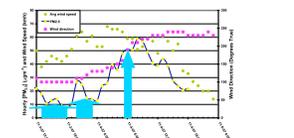


Figure 13. Wind Speed, Wave Height and $\text{PM}_{2.5}$ Measured at Sable Island 3-4 November, 2007.

Strong winds associated with Post Tropical Storm Noel generated significant sea salt aerosol, as seen in $\text{PM}_{2.5}$ measurements at Sable Island in November 2007. Wet removal as a sink is evident in the measurements during the hours rain was reported at Sable Island 3-4 Nov., 2007 (indicated by arrows in Figure 13). The default Monahan flux function resulted in an over-prediction of $\text{PM}_{2.5}$ compared to measurements at Sable Island in this case (Figure 14); Surface $\text{PM}_{2.5}$ concentrations resultant from using the "Monahan Surf" hybrid parameterization were under-predicted compared to measured $\text{PM}_{2.5}$ at Sable Island in this case.

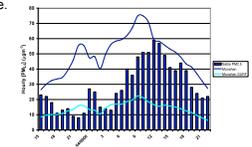


Figure 14. Measured versus Predicted $\text{PM}_{2.5}$ at Sable Island, 3-4 November, 2007